

A Sponge-based Moving Bed Bioreactor for Micropollutant Removal from Municipal Wastewater

by

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A thesis submitted in partial fulfilment
of the requirements for the degree of
Master of Engineering

University of Technology, Sydney
Faculty of Engineering and Information Technology

March, 2014

CERTIFICATE OF ORIGINAL AUTHORSHIP

I certify that the work in this thesis has not previously been submitted for a degree nor has it been submitted as part of requirements for a degree except as fully acknowledged within the text.

I also certify that the thesis has been written by me. Any help that I have received in my research work and the preparation of the thesis itself has been acknowledged. In addition, I certify that all information sources and literature used are indicated in the thesis.

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Acknowledgements

This thesis would not have been possible without the help and support of the outstanding individuals I met in the course of my research study. Now it is my turn to acknowledge them all.

First, and foremost, I would like to express the deepest gratitude to my supervisors Dr. Wenshan Guo and Prof. Huu Hao Ngo. I was amazingly fortunate to have the opportunity to be one of their research students. I enjoyed the great benefits of their invaluable suggestions, excellent guidance and endless inspiration. They deserve much of the credit for this thesis as well as my other outcomes.

I would like to extend my particular gratitude to A/Prof. Long Nghiem and Dr. Faisal Hai from the University of Wollongong for their valued feedback about my experiments and academic writing. Dr. Jinguo Kang is indebted for his help in the GC-MS analysis of micropollutants. Kaushalya Wijekoon is thanked for her assistance with the freeze drying of sludge samples.

I wish to thank Faculty of Engineering and Information Technology for providing a fantastic atmosphere for doing research. I am particularly thankful to Mohammed Johir who was being kind, generous and helpful throughout my experiments. Lijuan Deng is acknowledged for her assistance in some of my experimental analysis. My appreciation also goes to my other dear colleagues, Zhuo, Anwar, Zuthi, Atefeh, Hang, Bandita and Thanh, for their generous help in various aspects of this research.

Last but not least, I would also like to give my heartfelt thanks to my parents for their faith their encouragement in all of my pursuits. My sincere appreciation is also due to my friends who were always there to give unconditional support. I greatly value the friendship with them.

Table of Contents

Title Page	i
Certificate of Original Authorship	ii
Acknowledgement	iii
Table of Contents	iv
List of Figures	viii
List of Tables	x
List of Abbreviations	xiii
List of Symbols	xv
Abstract	xvi
Chapter 1 Introduction	1-1
1.1 Background	1-2
1.2 Statement on current issues regarding micropollutant removal	1-5
1.3 Objectives of the research	1-6
1.4 Outline of the thesis	1-7
Chapter 2 Literature Review	2-1
2.1 Introduction	2-2
2.2 Occurrence of micropollutants in the aquatic environment	2-2
2.2.1 Occurrence of micropollutants in WWTPs	2-2
2.2.2 Occurrence of micropollutants in surface water	2-5
2.2.3 Occurrence of micropollutants in groundwater	2-7
2.2.4 Occurrence of micropollutants in drinking water	2-7
2.3 Fate and removal of micropollutants in WWTPs	2-10
2.3.1 Fate of micropollutants in WWTPs	2-10
2.3.2 Overall removal of micropollutants in conventional WWTPs	2-13

2.3.3 Factors governing the fate of micropollutants in WWTPs	2-15
2.4 Overview of treatment alternatives for micropollutant removal	2-20
2.4.1 Coagulation–flocculation	2-20
2.4.2 Activated carbon adsorption	2-23
2.4.3 Ozonation and advanced oxidation processes (AOPs)	2-26
2.4.4 Membrane processes	2-30
2.4.5 Membrane bioreactor	2-33
2.5 Moving bed bioreactors (MBBRs)	2-38
2.5.1 Description of the MBBRs	2-38
2.5.2 Attached-growth carriers for MBBRs	2-39
2.5.3 MBBR applications in wastewater treatment	2-42
2.5.4 Micropollutant removal during MBBR processes	2-46
2.6 Conclusion	2-48

Chapter 3 Experimental Investigation 3-1

3.1 Introduction	3-2
3.2 Materials	3-2
3.2.1 Synthetic wastewater	3-2
3.2.2 Selected micropollutants	3-2
3.2.3 Sponge (polyurethane foam)	3-3
3.3 Experimental set-up and operation protocol	3-9
3.3.1 Batch experiments	3-9
3.3.2 MBBR system	3-9
3.3.3 MB-SMBR system	3-12
3.4 Analytical methods	3-14
3.4.1 Organic matter, nutrients, pH and DO	3-14
3.4.2 MLSS and MLVSS	3-14
3.4.3 Micropollutant	3-16
3.4.4 Fouling resistance	3-17

Chapter 4 Short-term Micropollutant Removal in Batch Experiments	4-1
4.1 Introduction	4-2
4.2 Organic and nutrients removal	4-3
4.3 Removal of selected micropollutants	4-3
4.4 Effects of sponge filling ratios on micropollutant removal	4-9
4.5 Conclusions	4-10
 Chapter 5 Removal of Micropollutants by a Moving Bed Bioreactor (MBBR) System	 5-1
5.1 Introduction	5-2
5.2 Organic matter and nutrient removal	5-3
5.3 Removal of selected micropollutants	5-8
5.4 Fate of micropollutants in the MBBR and application of mass balance	5-11
5.5 Comparison between the MBBR and other techniques for micropollutant removal	5-16
5.6 Conclusions	5-20
 Chapter 6 Removal of Micropollutants by a Moving Bed-Submerged Membrane Bioreactor (MB-SMBR) System	 6-1
6.1 Introduction	6-2
6.2 Organic and nutrient removal	6-3
6.3 Removal of selected micropollutants	6-8
6.4 Membrane fouling analysis	6-10
6.5 Conclusions	6-15
 Chapter 7 Conclusions and Recommendations	 7-1
7.1 Conclusions	7-2
7.2 Recommendations for future research	7-4

References	R-1
Publications Related to This Research	P-1

List of Figures

- Figure 1.1 Pathways for the introduction of micropollutants into the aquatic environment
- Figure 2.1 Average concentrations (on logarithmic Y axis) reported for the selected micropollutants in WWPT influents and effluents
- Figure 2.2 Maximum occurrence concentrations of some abundant micropollutants in drinking water (Benotti et al., 2008; Huerta-Fontela et al., 2011; Kleywegt et al. 2011; Vulliet et al., 2011; Wang et al., 2011)
- Figure 2.3 Sorption of micropollutants during biological treatment processes
- Figure 2.4 Removals of the selected micropollutants in WWTPs (data from Figure 2.1; negative removals not included). X-axis displays the selected compounds and their mean removal efficiencies and standard deviations (in the brackets). Error bars represent the standard deviations of the data
- Figure 2.5 Diagrams of aerobic and anaerobic or anoxic systems for MBBR technology
- Figure 2.6 Kaldnes type K1, K2 and K3 (from left to right) biofilm carriers (Rusten et al., 2006)
- Figure 2.7 Bioplastic-based moving bed biofilm carrier (virgin carrier, A) and (carrier with biofilm, B) (Accinelli et al., 2012)
- Figure 2.8 PVA gel beads (A) and Environmental scanning electron micrograph of the structure on the surface of an PVA gel bead (B, Rouse et al., 2005)
- Figure 2.9 Polyurethane sponge cubes with attached-growth biomass
- Figure 2.10 Typical process flow schemes for different MBBR applications
- Figure 3.1 The attached-growth carriers (sponge cubes) used in this study
- Figure 3.2 On-site photo and schematic diagram of the MBBR
- Figure 3.3 Schematic diagram of solvent extraction process of sludge samples
- Figure 3.4 On-site photo and schematic diagram of the MBBR-SMBR system
- Figure 3.5 The analytical instruments used in this study, including Analytikjena Multi N/C 2000 (A), Hach HQ 40d Portable Meter (B), Photometer NOVA 60 A Spectroquant (C), and Horiba OM-51 Portable Dissolved Oxygen Meter

- Figure 3.6 Schematic diagram of SPE process for GC-MS analysis of micro-pollutants
- Figure 3.7 Schematic diagram of processes for SMP and EPS extraction
- Figure 3.8 Relative hydrophobicity test (A) and microscopic analysis (B)
- Figure 5.1 DOC removal in the MBBR (aeration rate: 4 L/min; DO: 5.5–6.5 mg/L; HRT: 24 h; day 20 is the start of micropollutants addition; MPs: micropollutants)
- Figure 5.2 COD removal in the MBBR (aeration rate: 4 L/min; DO: 5.5–6.5 mg/L; HRT: 24 h; day 20 is the start of micropollutants addition; MPs: micropollutants)
- Figure 5.3 $\text{NH}_4\text{-N}$ removal in the MBBR (aeration rate: 4 L/min; DO: 5.5–6.5 mg/L; HRT: 24 h; day 20 is the start of micropollutants addition; MPs: micropollutants)
- Figure 5.4 TN removal in the MBBR (aeration rate: 4 L/min; DO = 5.5–6.5 mg/L; HRT: 24h; day 20 is the start of micropollutants addition; MPs: micropollutants)
- Figure 5.5 $\text{PO}_4\text{-P}$ removal in the MBBR (aeration rate: 4 L/min; DO: 5.5–6.5 mg/L; HRT: 24 h; day 20 is the start of micropollutants addition; MPs: micropollutants)
- Figure 5.6 Variation of attached biosolids and biomass concentrations in the MBBR
- Figure 5.7 Microscopic view of biomass growth within the sponge (A) virgin sponge; (B) sponge at the early stage; (C) sponge at the late stage
- Figure 5.8 Removal efficiency of micropollutants during the MBBR treatment. (An error bar represents the standard deviation of 25 measurements over 150 days; aeration rate: 4 L/min; DO: 5.5–6.5 mg/L; HRT: 24 h)
- Figure 5.9 Average concentrations of micropollutants on the suspended and attached biosolids (An error bar represents the standard deviation of the sample measurements on Day 70 and 100; aeration rate: 4 L/min; DO: 5.5–6.5 mg/L; HRT: 24h)
- Figure 5.10 Fate of the studied micropollutants in the MBBR system (aeration rate: 4L/min; DO: 5.5–6.5 mg/L; HRT: 24 h)
- Figure 6.1 DOC removal in the MB-SMBR system (MBBR conditions: aeration

- rate = 4.5 L/min, DO = 5.5–6.5 mg/L; HRT = 24h; SMBR conditions: aeration rate = 6 L/min, DO = 6.4–7.6 mg/L; filtration rate = 8.83 L/m²·h; HRT = 6h; SRT = ∞)
- Figure 6.2 COD removal in the MB-SMBR system (MBBR conditions: aeration rate = 4.5 L/min, DO = 5.5–6.5 mg/L; HRT = 24h; SMBR conditions: aeration rate = 6 L/min, DO = 6.4–7.6 mg/L; filtration rate = 8.83 L/m²·h; HRT = 6h; SRT = ∞)
- Figure 6.3 NH₄-N removal in the MB-SMBR system (MBBR conditions: aeration rate = 4.5 L/min, DO = 5.5–6.5 mg/L; HRT = 24h; SMBR conditions: aeration rate = 6 L/min, DO = 6.4–7.6 mg/L; filtration rate = 8.83 L/m²·h; HRT = 6h; SRT = ∞)
- Figure 6.4 TN removal in the MB-SMBR system (MBBR conditions: aeration rate = 4.5 L/min, DO = 5.5–6.5 mg/L; HRT = 24h; SMBR conditions: aeration rate = 6 L/min, DO = 6.4–7.6 mg/L; filtration rate = 8.83 L/m²·h; HRT = 6h; SRT = ∞)
- Figure 6.5 PO₄-P removal in the MB-SMBR system (MBBR conditions: aeration rate = 4.5 L/min, DO = 5.5–6.5 mg/L; HRT = 24h; SMBR conditions: aeration rate = 6 L/min, DO = 6.4–7.6 mg/L; filtration rate = 8.83 L/m²·h; HRT = 6h; SRT = ∞)
- Figure 6.6 Variations of MLSS and MLVSS concentrations in the SMBR
- Figure 6.7 Nematodes in the SMBR
- Figure 6.8 Micropollutant removal in the MBBR and MB-SMBR. An error bar represents the standard deviation of 8 measurements over 89 days (MBBR conditions: aeration rate = 4.5 L/min, DO = 5.5–6.5 mg/L; HRT = 24h; SMBR conditions: aeration rate = 6 L/min, DO = 6.4–7.6 mg/L; filtration rate = 8.83 L/m²·h; HRT = 6h; SRT = ∞)
- Figure 6.9 TMP profile over the 89 days of MB-SMBR operation
- Figure 6.10 Overgrowth of filamentous bacteria observed at the final stage of the study
- Figure 6.11 The fouled membrane after the MB-SMBR operation (SMBR conditions: aeration rate = 6 L/min; DO = 6.4–7.6 mg/L; filtration rate = 8.83 L/m²·h; HRT = 6h; SRT = ∞)

List of Tables

Table 1.1	Sources of micropollutants in the aquatic environment
Table 2.1	Human excretion rates of some common pharmaceutical compounds in the aquatic environment. (adapted from Alder, Hirsch et al., 1999; Huschek et al., 2004; Jjemba, 2006; Ternes, 1998; and the range was selected according to Jjemba, 2006)
Table 2.2	Occurrence of some common micropollutants in surface waters in different countries
Table 2.3	Occurrence of some common micropollutants in groundwater in different countries
Table 2.4	Simple classification of micropollutants based on removal efficiency
Table 2.5	Removals of some common micropollutants during coagulation–flocculation
Table 2.6	Removals of some common micropollutants during adsorption process
Table 2.7	Removals of some common micropollutants during ozonation and AOPs
Table 2.8	Removals of some micropollutants during membrane processes
Table 2.9	Removals of some micropollutants during MBR processes
Table 2.10	Recent publications on MBBR applications
Table 2.11	Recent publications on MBBR-MBR applications
Table 3.1	Composition and concentration of the stock solution
Table 3.2	Physicochemical properties of the selected trace organics
Table 3.3	Operational conditions of the batch experiments
Table 4.1	Variations of organic and nutrients levels during the batch experiments (aeration rate: 1.5 L/min; DO= 5–6 mg/L)
Table 4.2	Variation of micropollutant concentrations (ng/L) during the batch experiments (aeration rate: 1.5 L/min; DO= 5–6 mg/L)
Table 5.1	Comparison of micropollutant removal efficiency (%) in the MBBR and in other biological treatment technologies
Table 5.2	Assessment of different treatment processes for micropollutants removal
Table 6.1	EPS and SMP concentrations at different TMPs (SMBR conditions:

aeration rate = 6 L/min; DO = 6.4–7.6 mg/L; filtration rate = 8.83
L/m²·h; HRT = 6h; SRT = ∞)

Table 6.2 Membrane resistances for different types of fouling

List of Abbreviations

AOP	Advanced oxidation process
BAC	Biological activated carbon
CAFO	Concentrated animal feeding operation
COD	Chemical oxygen demand
CAS	Conventional activate sludge
DBP	Di-butyl phthalate
DEET	N,N-diethyl-meta-toluamide
DEHP	Di(2-ethylhexyl) phthalate
DMP	Di-methyl phthalate
DO	Dissolved oxygen
DOM	Dissolved organic matter
EDC	Endocrine disrupting compound
EPS	Extracellular polymeric substance
GAC	Granule activated carbon
GC-MS	Gas chromatography-mass spectrometry
HRT	Hydraulic retention time
MBBR	Moving bed bioreactor
MBR	Membrane bioreactor
MB-SMBR	Moving bed-submerged membrane bioreactor
MF	Microfiltration
MLSS	Mixed liquor suspended solids
MLVSS	Mixed liquor volatile suspended solids
NF	Nanofiltration
NLR	Nitrogen loading rate
NOM	Natural organic matter
NSAID	Nonsteroidal anti-inflammatory drug
OLR	Organic loading rate
PAC	Powdered activated carbon
PCP	Personal care product
PNEC	Predicted no effect concentration
PPCP	Pharmaceutical and personal care product

PU	Polyurethane
PVA	Polyvinyl alcohol
PVDF	Polyvinylidene fluoride
RO	Reverse osmosis
SAnMBR	Submerged anaerobic membrane bioreactor
SBBGR	Sequencing batch biofilter granular reactor
SBF	Sponge biofilter
SBR	Sponge batch reactor
SMBR	Submerged membrane bioreactor
SMP	Soluble microbial products
SND	Simultaneous nitrification–denitrification
SPE	Solid phase extraction
SRT	Sludge retention time
TCEP	Tris(2-chloroethyl) phosphate
TCPP	Tris(1-chloro-2-propyl) phosphate
TMP	Transmembrane pressure
TN	Total nitrogen
TOC	Total organic carbon
TP	Total phosphorus
UF	Ultrafiltration
WWTP	Wastewater treatment plant

List of Symbols

$C_{s, a}$	Concentration of micropollutant on the attached biosolids ($\mu\text{g/g}$)
$C_{s, s}$	Concentration of micropollutant on the suspended biosolids ($\mu\text{g/g}$)
$C_{w, \text{eff}}$	Average effluent concentrations of micropollutants (ng/L)
$C_{w, \text{inf}}$	Average influent concentrations of micropollutants (ng/L)
J	Permeation flux ($\text{L/m}^2\cdot\text{h}$)
K_d/D	Solid-water distribution coefficient
K_H	Henry's law constant
K_{OW}	Octanol–water partition coefficient
L_{sol}	Load of micropollutant removed via sorption over 30 days (ng)
MLSS	Mixed liquor suspended biosolids concentration (g/L)
pK_a	Acid dissociation constant
ΔP_T	Transmembrane pressure (kPa)
Q	Flow rate of the MBBR (L/day)
R_c	Cake resistance formed by cake layer deposited over membrane surface (m^{-1})
R_f	Fouling resistance caused by pore plugging and/or solute adsorption onto the membrane pore and surface (m^{-1})
R_m	Intrinsic membrane resistance caused by membrane itself and permanent resistance (m^{-1})
ΔSS	Increased amount of attached biosolids over the study period (g)
T	Duration of the study period (day)
μ	Viscosity of the permeate (m^2/s)

Abstract

Over the past few decades, the frequent detection of micropollutants in the aquatic environment has raised particular health and environmental concerns. Wastewater treatment plants (WWTPs) serve as significant barriers to reduce the release of micropollutants. However, due to the diverse characteristics and low concentrations of micropollutants, WWTPs can only achieve variable and often inadequate removals, ranging from 12.5% to 100% for some frequently reported compounds.

This study investigated a sponge-based MBBR for its effectiveness in the elimination of various micropollutants, including pharmaceuticals and personal care products, steroid hormones, industrial chemicals and pesticides. A moving bed-submerged membrane bioreactor (MB-SMBR) was also evaluated in terms of micropollutant removal and membrane fouling.

During the batch experiments, non-acclimatized (virgin) sponge showed significant and rapid sorption of hydrophobic compounds. Acclimatized sponge could achieve much higher elimination of some acidic pharmaceutical compounds, such as acetaminophen, diclofenac, gemfibrozil, ibuprofen, ketoprofen, naproxen and salicylic acid. Carbamazepine, fenoprop and metronidazole were poorly removed during all the batch experiments.

The sponge-based MBBR was effective in removing organics and nutrients (except $\text{PO}_4\text{-P}$). Most of the selected micropollutants (16 out of 22) showed removals of higher than 70%. The poorly or moderately removed compounds included carbamazepine (25.9%), fenoprop (31.0%), diclofenac (45.7%), metronidazole (54.8%), ketoprofen (58.2%), and gemfibrozil (62.4%). The low biodegradability and/or polar property were two causes for the insufficient elimination. Overall, the effectiveness of the MBBR for micropollutant removal was comparable with those of other biological treatment processes, including activated sludge and membrane bioreactors (MBRs). Biodegradation was the major removal mechanisms for most compounds during the MBBR treatment. Sorption was only significant for the

refractory compounds, while the readily biodegradable compounds did not considerably accumulate on the biosolids.

With the incorporation of the SMBR, the whole system (MB-SMBR) could significantly reduce the effluent turbidity. However, the SMBR did not achieve much supplementary removal of organic, nutrients and micropollutants. The membrane fouling in the SMBR occurred to a minor extent during the first 84 days of operation, after which an abrupt TMP increase was observed. High EPS levels (16.24 mg/L) in the SMBR was a potential cause for the severe fouling. The overgrowth of filamentous bacteria could also be deemed a factor that accelerated the membrane fouling rate. The total membrane resistance was mainly attributed to the deposited cake layer (76.5%), followed by the pore blocking (12.0%), clean membrane resistance (10.5%) and irreversible fouling (1.0%).